Problems with accurate atomic lifetime measurements of multiply charged ions

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Abstract
A number of recent atomic lifetime measurements on multiply charged ions have reported
uncertainties lower than 1%. Such a level of accuracy challenges theory, which is a good
thing. However, a few lessons learned from earlier precision lifetime measurements on atoms
and singly charged ions suggest that one should remain cautious about the systematic errors of
experimental techniques.

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1. Introduction

Einstein *A* and *B* values describe the spontaneous and
stimulated absorption and emission of light by atoms;
oscillator strengths *f* and line strengths *S* can be converted
into each other and into *A* values, if the transition energies
are known. Quantum mechanics (QM) has been with us for
more than 80 years, relativistic quantum mechanics (Dirac
equation) for more than 70 years, quantum electrodynamics
(QED) for about 60 years. Nobel prizes have been awarded
for atomic physics work at the relative uncertainty level
of 10^{-12}–10^{-15}, and work continues that aims for even
higher precision. Surely everything worth knowing about
atoms is known with high precision, and we can turn away
from fundamental atomic physics in favour of applying the
knowledge elsewhere? Well, the atomic structure knowledge
is, in fact, patchy. There is very notable recent progress
in terms of the accuracy of *ab initio* calculations that
reduces the gap between calculations for a single-electron
system (or a single electron outside of nominally closed
atomic shells), for which very good calculations have been
achieved decades ago [1, 2]; these calculations have been
corroborated by experiment up to $Z = 92$ [3]. Evidently
the accuracy in the description of atomic systems with
more than one electron in the valence shell is much lower,
and accurate predictions have begun to emerge only rather
recently [4]. Atomic spectra, after all, relate to the differences
of atomic level energies, and when taking the differences of
large numbers (of electron binding energies, for example),
the relative uncertainty of the difference is much higher

than that of the original numbers. Notwithstanding laser
spectroscopy, with its much higher capabilities for spectral
resolution and frequency determination (mostly) in the visible
range of the electromagnetic spectrum and in observations
of atomic beams and carefully trapped particles, studies in
other spectral ranges and on multiply charged ions (because
of the light source properties) largely rely on classical
spectroscopy. There, a typical figure of merit would be
an accuracy of the wavelength determination at the 10^{-4}
level (astrophysical observations aiming at extrasolar planets
need higher precision), and at present theory cannot deliver
that for arbitrary ions. The theoretical understanding of
atomic structure is much further advanced than the practical
implementation of the QM of many-body systems into
computer codes. The QED contribution to atomic levels and
level differences nowadays is considered to be under better
control than the many-body QM part.

However, even poorer is the reliability of predictions
and data on transition probabilities, that is, on Einstein *A*
values. Here theory and experiment have similar difficulties.
Experiment usually determines the total decay rate *R* of an
atomic level *k* to all other (lower) levels *i*:

$$R_{\text{total}} = \Sigma(A_{ki}),$$

regularly in the form of a level mean life τ

$$\tau_k = 1/\Sigma(A_{ki}).$$

Only in the case of unbranched decays can the lifetime be
inverted to yield a single *A* value. Many precision lifetime

measurements in fact deal with such unbranched decays. Accurate branching fractions, however, are also a treasure trove of information on atomic structure physics [5], but presently they have their own limits on precision that are beyond the scope of this Comment. I will use a figure of merit of 1% as a marker for precision in this discussion on atomic level lifetimes. This level of precision has been a rarely met challenge for decades. New techniques (using a heavy-ion storage ring or an electron beam ion trap (EBIT)), specifically for highly charged ions, have broken through this 'barrier' recently, and problems have promptly surfaced that relate to theory on the one hand and to experiment on the other.

If either theory or experiment on a given topic are insufficiently developed, not much can be learned. It is for this reason that one-electron atomic systems have been pursued for which theoretical predictions are considered to be nearly perfect. Sometimes it has turned out to be advantageous experimentally to study a system with one electron outside closed shells (Li-, Na-, Cu-like ions), as in the example of the best measurement of the two-loop Lamb shift measured in the Li-like ion U^{89+} [6]; this, however, has required calculations to match the high measurement precision so that the data obtained on the three-electron ion could be interpreted in terms of the corresponding entities in single-electron U^{91+}. In this case, the indirect evidence gathered experimentally from a three-electron ion was still more accurate than any direct energy measurements of one-electron ions by almost an order of magnitude.

2. Historical notes

As an example, 35 years ago the Berlin group of H J Andrä applied laser spectroscopic techniques to fast atom and ion beams and reported precise measurements of the lifetime of the 6 p level of Ba II ($\pm$1%) [7] and, a few years later, of the 6s6p $^1P_1$ level of Ba I ($\pm$0.25%) [8]. However, there was no theory available to match this precision for several decades. The experimenters therefore turned to atomic systems (Li and Na atoms) [9, 10] for which theory did provide numbers. Surprisingly, theory was found to be off the mark—with the exception of the numerical Coulomb approximation, which was considered as being too simplistic to be possibly accurate. Other types of experiment, for example using laser-induced fluorescence in a stationary target [11], did not quite reach the small error bars of the Berlin experiment. Over the course of some 15 years, theory evolved and then cast doubts on the full quality of the early measurements. Eventually, refined atomic beam experiments [12–14], and also a very different approach using molecular states [15, 16], superseded the old data and corroborated the intervening better calculations. Interestingly, some of the earlier measurements with somewhat more conservative error estimates [10, 11] survived unbeaten.

The above laser spectroscopic measurements concerned atoms and singly charged ions with atomic levels of nanosecond lifetimes. Recently there have been measurements of millisecond level lifetimes in multiply charged ions that do not agree with modern theory, and the earlier experiment versus theory situation may be repeated. Although the working range is very different, maybe the interpretation of the new experiments can profit from experiences made with the old ones.

In common gas discharge light sources, atomic levels with nanosecond lifetimes dominate, although much-longer lived (metastable) levels play important roles for the excitation process and ionization balance in fluorescent tubes. In high pressure lamps, collisions broaden the emission lines, and, from a high-density plasma, characteristic line emission can be detected only from very short-lived ions. The much lower collision frequencies of low-density plasmas (tokamaks, etc. in the laboratory, or the solar corona) let us see the emission of much longer lived levels, because they are not quenched. Some designs of future frequency standards rely on very long-lived levels. Here the long level lifetime corresponds to a vanishingly small level width and thus a narrow spectral line width, but the actual lifetime value is of little interest. A practical interest in reasonably precise atomic level lifetimes exists for the industrial optimization of light sources as well as for the diagnostics of plasmas via collisional-radiative modelling. Highly precise lifetime data are mostly an intellectual challenge, checking on the validity of such models and thus on the underlying understanding of atomic structure and dynamics.

It is striking how poor an overall lifetime uncertainty of 1% looks in comparison to some wavelength determinations that have reached a level of 10^{-15}. There are several very simple reasons for this. The determination of the position of a spectral feature usually takes place within a small working range, the overall position (a very large number in transition frequency space) of which has been established by other means. Therefore only a minute 'correction' needs to be added, for which on its own the requirement for accuracy is but moderate. In particular, the position of a symmetric spectral line—relative to a well-established calibration curve—often requires only a few measurement points and a fit of an almost arbitrary symmetric curve shape to the data points. In contrast, atomic lifetime measurements carry the full uncertainty (in addition, for example, to a reference clock). If the lifetime is to be determined from a linewidth, a Lorentzian, a Gaussian, or a Voigt profile has to be fitted to a much more extensive data sample, most data points of which are far out in the line wings and therefore near to the background level. If the level lifetime is measured in the form of an exponential decay curve, no symmetry argument is available to reduce the uncertainty of the fit. Exponential components are similar to each other, and the fitting of several such components (from blends or from cascade repopulation) is a nonlinear process and compounds the uncertainty. The reference timescale and the signal-to-noise ratio of a given detector are of utmost importance as well. Furthermore, good detectors are not available in all spectral ranges.

Precise lifetime measurements on atoms and some on singly charged ions usually employ laser excitation (see [17]). Laser excitation has been applied in a variety of experiments, such as metastable of rare gases (pre-excited in a discharge or in a collision cell), fast atomic beams (produced from fast ion beams by electron capture in a dilute gas), and excited gas leaking from a discharge vessel. In this way, the rates of a large number of electric dipole (E1) transitions have been measured with precision in the range of a few percent [18]. However, with the exception of the alkali atoms, theory does not compete for E1-dominated level lifetimes of complex

atoms at the accuracy level of below 1% and hardly any experimental data challenge theory at this level.

Lasers are generally not suited to reach excited levels (from the ground state or a low-lying metastable level) in multiply charged ions, nor to produce and excite in a selective way multiply charged ions from a neutral target. For several decades, beam-foil spectroscopy (BFS) [19] was the foremost technique to be employed for lifetime measurements of atomic levels (multiply excited levels, levels of multiply charged ions) that laser excitation could not reach. In this technique, a fast ion beam of well-specified energy is passed through a very thin foil, with a high yield of excitation of the projectile electrons in the energetic interaction with the electrons of the solid target. However, this interaction also causes a small energy loss and some angular straggling, both of them sufficiently large and too uncertain to allow high accuracy. The highest lifetime precision (0.27%) of any classical beam-foil experiment [20] has been reached when using a time reference within the actual foil-excited ion beam, exploiting quantum beats (the result of coherent excitation) from a well calculated and thus precisely known fine structure interval in neutral helium. The quantum beats were observed concurrently with the light of primary interest, but through a filter for a different wavelength. Thus, the decay signal and the atomic time reference were recorded in parallel. The usually much larger fine structure intervals in multiply charged ions mean very high quantum beat frequencies with poor contrast in observations, and therefore they preclude the extension of that technique. Only very few BFS lifetime measurements have reached an uncertainty of less than 1%, and much of the limitation owes to the problem of determining the energy loss of the fast ions in the exciter foil whose precise thickness is difficult to establish and which also changes under irradiation.

An interesting precision lifetime measurement ($\pm 0.4\%$) on a singly charged ion has been performed by using monochromatized synchrotron radiation to ionize Ar and to selectively excite its first level above the ground state, the $3s3p^6\ ^2S_{1/2}$ level [21] (and similarly in Kr II and Xe II). Earlier beam-foil experiments had reached uncertainties no better than 5–10% on such levels in the same ion or in neighbouring ions along the isoelectronic sequence. Among the results of earlier calculations, some were up to five times larger than the smallest—for a given element. A new calculation (alas, presented after the experimental fact) [22] has found reasonable agreement with the experimental lifetime value for Ar II, but has not reached any further along the isoelectronic sequence. Beam-foil data are available for a number of elements of this sequence, as well as calculations (see [23–25] and references therein). However, none of the other measurements are stated with an accuracy of just a few percent.

For most ions with several electrons in the valence shell the practical determination of experimental lifetimes is hampered by the occurrence of cascade repopulation of a level of interest by the decays of other levels that are being excited as well. Selective laser excitation of foil-excited ions has been demonstrated (the ion–foil interaction was employed to first make ions of the proper charge state) [26], but the energy distribution in a 'monoenergetic' ion beam is much wider than the band width of a good laser, and therefore

only very few ions actually interact with the laser light on resonance at a given time. The cascades often have lifetimes of the same order of magnitude as the levels of interest, but multi-exponential fitting algorithms can analyse reliably only those decay curves in which the components differ substantially in time constant. The arbitrarily normalized direct cascades (ANDC) technique [27] in which not only the decay of the level of interest, but also all cascades feeding the very same level are measured, may well be able to avoid systematic errors of the order of 50% or more. Unfortunately, not many cases are known in which all such cascades can be measured in practice.

3. Transition types

BFS has largely been limited to E1 transitions (and some M1 and M2 transitions in more highly charged ions) with level lifetimes in the range from a few picoseconds to hundreds of nanoseconds. E1 transition rates yield information on atomic wave functions that supplements the insight gained from atomic energy levels alone. The E1 transition rate depends on the transition energy and an extra power of r through the electric dipole operator er. Transitions between fine structure levels of a given term (E1-*forbidden* transitions, mostly M1 and E2 transitions) are supposedly insensitive to this, as they connect levels with similar radial wave functions. However, complex wave functions as well as relativistic effects in highly charged ions modify this simple picture. In very highly charged ions, M1 and E2 transition rates compete with E1 transition rates; in ions of low to moderate charge states, however, their rates are lower by several orders of magnitude, and the corresponding atomic level lifetimes are in the millisecond to second range. Their measurement necessitates the application of ion trapping techniques. Otherwise ions at thermal energies or higher would escape from the observation volume before radiative decay has a chance to take place.

Conventional ion traps (of Kingdon, Penning, or Paul type) have all been used for atomic lifetime measurements (for reviews, see [28–30]), but have not yielded atomic lifetime results with accuracies of 1%. This has been achieved only with a heavy-ion storage ring or an EBIT. Both types of device are covered in some detail in the above reviews. The heavy-ion storage ring can be seen as an extension of the beam-foil scheme: after excitation (in the injector accelerator or by a laser), the ion beam travels in a roughly circular vacuum vessel and passes the detection zone (a photon detector or an electron target followed by a particle detector) over and over again. The actual ion beam energy is of little importance; since there is no displacement of mechanical components necessary in order to measure a decay curve, only electronic timing and a determination of the ion loss rate of the stored ion beam are required.

4. Relativistic M1 transitions

An accurate lifetime measurement of this type was first achieved at the Heidelberg heavy-ion test storage ring (TSR). In a stored beam of He-like ions (C^{4+}, N^{5+}) the presence of excited ions in the $1s2s\ ^3S_1$ level was probed by merging a section of the ion beam with a 'cold' electron beam at a

precisely determined different velocity. If the electron beam energy in the rest frame of the fast ions matches a resonance for dielectronic recombination, an electron can be attached to an ion, by exciting one of the ion's electrons. From this doubly excited state, the ion can either autoionize and thus return to its previous condition, or stabilize radiatively and thus change its charge state, which is easy to detect by catching the ion on its now different trajectory in the next magnetic dipole field of the beam guidance system. Excitation and probing involve such small cross sections that the stored ion beam is practically undisturbed. The Heidelberg lifetime measurements on C^{4+} and N^{5+} [31], later extended to B^{3+} ions, have reached accuracies of as good as 0.2%, for level lifetimes in the range of many milliseconds. The lifetime data test the calculations of a relativistic operator, which enables a magnetic dipole decay of the lowest triplet level in two-electron ions, one of the simplest atomic systems and thus a particularly fundamental case in atomic structure theory. A later extension to Be^{2+} ions made use of the larger ion loss rate of excited ions versus ground state ions exposed to collisions with the residual gas in the excellent high vacuum of a storage ring [32], but the 1.8 s lifetime could not be determined to better than about 2%.

The same M1 transition, but in He-like ions of elements N through S [33–35] (lifetimes from a few milliseconds to less than one microsecond), has been studied at the Livermore EBIT. In these measurements, an electron beam breeds ions of the desired charge state inside a Penning trap. When the electron beam is switched off, the ions remain trapped [36], and their delayed x-ray emission can be recorded in order to accumulate a decay curve. Owing to the excellent vacuum in a cryogenic EBIT, the ion loss rates are much lower in an EBIT than in conventional ultra-high vacuum (UHV) Penning traps; control of the vacuum is indeed one of the key parameters in obtaining high accuracy in atomic lifetime measurements using storage rings or EBITs [37]. The lifetime measurements on x-ray transitions owe part of their quality also to the fact that in the x-ray range detectors can be operated with almost perfect noise rejection.

The Heidelberg TSR and Livermore EBIT lifetime data on He-like ions are part of the longest isoelectronic sequence studied for a particular level lifetime, the results for neutral He to Xe^{52+} spanning about 15 orders of magnitude. The above results are by far the most accurate among them. For Ar and beyond, beam-foil and recoil ion beam measurements have typically reached uncertainties of a few percent. For He and the light ions, the atomic lifetimes are so long that presently no experimental technique can render them with anywhere near the desired accuracy. The accurate lifetime results for O^{6+}, Ne^{8+} and S^{14+} ($\pm 0.5\%$) agree well with nonrelativistic calculations after a leading-term relativistic correction of the transition rate as well as with fully relativistic calculations that presently set the standard [38–40]. The mutual agreement of these calculations (of a relatively simple atomic system) with each other and with the Heidelberg heavy-ion storage ring and Livermore EBIT lifetime data (within the latters' 0.5% measurement uncertainty) can also be interpreted as a corroboration of the measurement techniques.

5. Spin-changing E1 (intercombination) transitions

At both TSR and the Livermore EBIT, lifetime measurements have also addressed more complex ions with transitions accessible with use of photomultiplier tubes (PMT). With about $60 \, \mu A$ of C^{2+} ions in the storage ring, the PMT signal rate of the intercombination decay of this ion was about 10 counts per second [41], whereas the dark count rate of the solar-blind PMT was less than 1 count per second. The maximum operating cycle frequency of TSR is about 5 Hz, so that the shortest storage time intervals are about 200 ms. The signal count rate relates to an atomic level lifetime of about 10 ms; the accumulated decay curves reached a peak-to-tail ratio of better than 200, corresponding to more than 5 lifetimes before the signal reached the background level. Such a measurement, after accounting for systematic error caused by ion loss estimates of the possible influence of the magnetic dipole fields of the ion-beam guidance magnets, was quoted with an accuracy of 0.14%, the most accurate lifetime measurement on any multiply charged ion at the time. A relativistic configuration interaction (RCI) calculation of this case, involving a massive effort of including some 200 000 wave functions [42] is quoted with an intrinsically justified uncertainty of as little as 0.5% and disagrees with the experiment by slightly more than the combined error bars. This small high-level discrepancy is worth speculating about, whether there are unrecognized shortcomings in the calculation or in the experiment. There are additional less accurate experimental data for the same intercombination transition in Be-like ions B, N and O, and they seem to be isoelectronically consistent (see [43]), but no similar massive calculational effort has been spent on any of these other ions.

Intercombination transitions in several other atomic systems have been measured similarly at TSR, with some small deviations from calculations. It is an open question how to corroborate further the validity of the experimental technique (except for the PMT, the same basics apply as in the successful measurements on He-like ions) or whether multi-electron ions are subject to extra influences in the storage ring environment. Better calculations would be most welcome, too.

6. E1-forbidden transitions

Electric-dipole-forbidden transitions within the ground configurations of multiply charged ions, usually M1 (often with small E2 admixtures), are the staple of plasma diagnostics in both astrophysical and terrestrial plasmas. The associated level lifetimes in a number of ions have been measured at TSR and at several EBITs. Systematic work at Livermore has indicated shortcomings of earlier attempts elsewhere that probably related to vacuum conditions under insufficient control, while the single Oxford EBIT lifetime measurement was corroborated. For a while, the Livermore EBIT lifetime measurements on transitions in the visible appeared limited to an accuracy of about 2%, which was reached on B-like ions of Ar and neighbouring elements. Theoretical predictions fell within 1%, but only after replacing whatever calculated transition energy ΔE by the experimentally determined one, and combining this

with a line strength S that was practically identical to the basic single-configuration limit value. In a sense, theory had the predictive value of indicating the correct principle of how to combine S and ΔE (by different powers of ΔE for different transition types), but the practical calculations had little quantitative predictive value. Only rather recently the first *ab initio* calculations of certain M1 dominated level lifetimes have matched experimental data quality. For example, the multi-reference Møller–Plesset calculations by Vilkas and Ishikawa [44] have matched the (0.6%) Livermore EBIT lifetime measurement on (Al-like) Fe XIV [45]. The Livermore lifetime measurement has recently been corroborated by an even more precise one at the newer Heidelberg EBIT [46], and the comparable, but more difficult to measure corresponding lifetime in Fe X has also been measured there, the result being in agreement with theory and with the (better than 1%) extrapolation of measurements on isoelectronic ions at the Heidelberg heavy-ion storage ring [47–49].

The aforementioned B-like ion of Ar with its M1 transition that is rather well suited to precision measurements (Ar is easy to introduce into an EBIT, the wavelength of about 442 nm is almost ideal for detection by PMTs) has also been studied at the Heidelberg EBIT, with an error estimate of less than 0.1% [50, 51]. This is a remarkable achievement, but what does the comparison of such an accurate result to theory mean? At face value, the measurement disagrees with all available calculational results, by several (experimental) standard deviations, and the predictions themselves scatter by more than that (and lie on one side). However, as pointed out above, most calculations carry no uncertainty estimate, and therefore they are outright useless for any such comparison of quality. Most of them furthermore rely on experimental transition energies—again, there is no particular predictive value in most of these calculations. The Heidelberg group had excellent theory support of their own; their own calculation represents the state of the art. The QM result for the transition energy would be corrected for QED contributions, which have a 0.4% effect on the transition rate. Alternatively, one can return to basic principles, combining the experimental transition energy (where nature takes care of the QED level shift) with the line strength S. Configuration mixing might affect S, but near the low end of the isoelectronic sequence, this should be a minor problem. Alternatively, the lifetime measurement might be interpreted as a measure of S.

However, there is a QED correction for the anomalous magnetic moment of the electron (EAMM, or (g-2)) that has been mentioned in [50] as being easily derived, but which had been largely overlooked before. This EAMM correction acts as a multiplier $(1 + 2\alpha/\pi)$ on the transition rate and affects the rate and thus also the level lifetime by about 0.45%. The Heidelberg experiment on Ar^{13+} represents the first test of such a QED transition operator correction in an ordinary atomic system. In combination with the experimental transition energy value, which has also been determined to extreme accuracy at the Heidelberg EBIT [52], the lifetime measurement could serve as a valid test of this particular QED correction—if the experimental accuracy of the lifetime determination could somehow be corroborated, perhaps by other measurements or at least measurements in other places.

In the present situation, without independent experimental cross checks, it has been suggested [53] to consider the otherwise unexplained discrepancy as a measure of systematic error of the measurement—which would be larger than conceded by the Heidelberg experiment team, who have run a wide range of systematic error checks, of course.

In a second lifetime measurement at the Heidelberg EBIT, the Al-like ion Fe^{13+} (spectrum Fe XIV) was studied [46]. In a way this ion is similar in structure to the B-like ions, just with principal quantum number $n = 3$ instead of $n = 2$. The disagreement with theory came out as 1%, or about 10 standard deviations, and also on the long lifetime side of prediction. For Fe XIV there are four other measurements, two using electrostatic ion traps [54, 55], one using a heavy-ion storage ring [56], and one using the EBIT at Livermore [45]. All have reported lifetime results on the long side of theory. Uncommonly, the result obtained at a heavy-ion storage ring carried the largest error bar (and deviation of the mean from prediction) of the sample, although in many other experiments the heavy-ion storage ring results were to be most accurate. This observation may yield a clue (see below) to a possible (much smaller) systematic error in the corresponding EBIT measurements.

7. Cascade story

In BFS, atomic lifetimes that appear too long usually suffer from cascade repopulation after non-selective excitation. If one assumes for the beam-foil error 10% of the atomic lifetime sought, a cascade contribution that is weaker by two orders of magnitude would be difficult to observe directly, but might cause a systematic error of about 0.1%. Evidently, this is a magnitude that matters in accurate lifetime measurements. Such a cascade repopulation effect has been discussed in the context of EBIT lifetime measurements at Livermore [45], but based on collisional excitation calculations and in comparison to a 0.7% lifetime measurement then reported, it was deemed a minor error contribution. However, the Heidelberg EBIT measurements [46, 50, 51] cite an error that is smaller by an order of magnitude, and then the possibility of a cascade contribution might need to be reconsidered. Beyond the usual cascades from high-lying bound states, Nicolaides [57] has pointed out low-lying continuum states as a reservoir of cascades after radiative recombination, which should be more prominent in highly charged ions. In a storage ring, the fast ions experience the magnetic dipole fields of the beam guidance magnets also as motional electric fields. The populations of high-lying levels beyond $n \approx 30$ or 40 are expected to be quenched over time by such fields, so that these continuum states should not contribute much to the actual decay curves. However, if the initial ion excitation is by ion–foil interaction, the overall reservoir may still be sufficient for a notable influence.

The peculiar cascade situation in Al-like ions that has become apparent at the heavy-ion storage ring concerns a 3d level of millisecond lifetime feeding a 3p level of millisecond lifetime while practically all other levels have picosecond and nanosecond lifetimes. Similar cascades appear in Si- and S-like ions [58], but no comparable cascades have yet been identified in B-, Mg-, P-, or Cl-like ions. Nevertheless, the

highly precise lifetime measurements on B-like Ar XIV (at the Heidelberg EBIT) [50, 51] differ systematically from the available predictions. The discrepancy is smaller than the case of (Al-like) Fe XIV; once the latter was corrected (approximately) for the aforementioned specific cascade, both Heidelberg EBIT measurements would show about the same level of disagreement with theory. So far the two Heidelberg EBIT measurements are the only ones on M1 transitions that claim an uncertainty as small as 0.1%. The Livermore EBIT result [45] is the next precise, and it also deviates from the present state of prediction by more than one standard deviation. However, with the assumed cascade correction, the discrepancy with theory would disappear. It is of much interest to find out whether the remaining (Heidelberg) disagreement of the order of 0.3–0.5% results from an unrecognized systematic error beyond the cascade problem which is traceable to specific long-lived levels, or whether this is a glimpse at new physics. (We note that for the aforementioned other highly precise measurement, a storage ring experiment on the intercombination transition in Be-like C III [41], no QED correction to a transition operator applies, and the actual deviation from highly developed theory [42] has the opposite sign.) The Heidelberg EBIT measurements apply to M1 transitions. QED corrections corresponding to the one for M1 transitions may be applicable to other E1-forbidden transition rates, but we have not found any theoretical presentation of this topic or whether there are further corrections that might depend, for example, on the nuclear charge or on the principal quantum number n.

8. Note of caution

Once the QED correction to the M1 transition operator is fully understood and the (about) 0.5% discrepancy between the EBIT results and theory clarified, will then all lifetime-related problems be overcome, at least down to an accuracy of 0.5%? Not quite. Take the example of hyperfine quenching. The effect has been conceptualized and understood for nearly 80 years. In He-like ions, the agreement of theory and experiment seemed satisfactory. In Be- and Mg-like ions, however, recent calculations by Cheng *et al* [60] have arrived at predictions that differ by up to 80% from earlier ones, and by about 20% from a very recent experiment on Be-like ^{47}Ti^{18+} ions [59]. The experiment employed a heavy-ion storage ring and dielectronic recombination, a highly selective detection process; the uncertainty of the result was given as 5%. This is some way from the 1% guideline of this Comment, but it underlines how difficult it will be to ascertain accuracy in certain cases, even some that are considered to be relatively simple. And in this case, theory does not even agree with the experimental result.

Is there a path that promises the reduction of many systematic errors, as has been achieved by laser excitation of fast ion beams decades ago? Indeed, the same principle, but adjusted to multiply charged ions, should do: selective excitation (or at least selective modulation of the population) of levels can suppress the cascade problem. Selective excitation of resonance levels in highly charged ions of various elements has been achieved by the Heidelberg EBIT group at the FLASH facility (Free-Electron Laser in

Hamburg), where intense EUV light sufficed to selectively excite the resonance transitions in separately prepared Li-like Fe ions [61] and in similar ions of heavier elements. It should be rather straightforward to turn this approach into a time-resolved observation that would yield the resonance level lifetimes (which are in the range of dozens of picoseconds). Without cascades and without much of a background problem, the measurement promises high accuracy for the lifetime, especially as corrections for ion losses are unimportant on this timescale. However, lifetime measurements in atomic physics continue to provide surprises, at the high-accuracy level as well as in unexpected corners.

References

[1] Kim Y-K, Baik D H, Indelicato P and Desclaux J P 1991 *Phys. Rev.* A **44** 148
[2] Blundell S A 1993 *Phys. Rev.* A **47** 1790
[3] Träbert E, Beiersdorfer P and Chen H 2004 *Phys. Rev.* A **70** 032506
[4] Vilkas M J and Ishikawa Y 2005 *Phys. Rev.* A **72** 032512
[5] Curtis L J, Federman S R, Torok S, Brown M, Cheng S, Irving R E and Schectman R M 2007 *Phys. Scr.* **75** C1
[6] Beiersdorfer P, Chen H, Thorn D B and Träbert E 2005 *Phys. Rev. Lett.* **95** 233003
[7] Andrä H J, Gaupp A and Wittmann W 1973 *Phys. Rev. Lett.* **31** 501
[8] Andrä H J 1976 *Beam Foil Spectroscopy* ed I A Sellin and D J Pegg (New York: Plenum) p 835
[9] Gaupp A, Kuske P and Andrä H J 1982 *Phys. Rev.* A **26** 3351
[10] Schulze-Hagenest D 1979 *PhD Thesis* University of Kaiserslautern unpublished
[11] Carlsson J 1988 *Z Phys.* D **9** 147
[12] Volz U, Majerus M, Liebel H, Schmitt A and Schmoranzer H 1996 *Phys. Rev. Lett.* **76** 2862
[13] Volz U and Schmoranzer H 1996 *Phys. Scr.* T **65** 48
[14] Oates C W, Vogel K R and Hall J L 1996 *Phys. Rev. Lett.* **76** 2866
[15] McAlexander W I, Abraham E R I, Ritchie N W M, Williams C J, Stoof H T C and Hulet R G 1995 *Phys. Rev.* A **51** R871
[16] McAlexander W I, Abraham E R I and Hulet R G 1996 *Phys. Rev.* A **54** R5
[17] Scholl T J, Holt R A, Masterman D, Rivest R C, Rosner S D and Sharikova A 2002 *Can. J. Phys.* **80** 1621
[18] Den Hartog E A, Lawler J E, Sneden C and Cowan J J 2003 *Astrophys. J. Suppl. Ser.* **148** 543
[19] Träbert E 1997 in *Accelerator-based Atomic Physics Techniques and Applications* ed S M Shafroth and J C Austin (Washington: AIP) p 567
[20] Astner G, Curtis L J, Liljeby L, Mannervik S and Martinson I 1976 *Z. Phys.* A **279** 1
[21] Lauer S, Liebel H, Vollweiler F, Schmoranzer H, Lagutin B M, Demekhin Ph V, Petrov I D and Sukhorukov V L 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** 2015
[22] Saha B and Fritzsche S 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** 1161
[23] Träbert E 1996 *J. Phys. B: At. Mol. Opt. Phys.* **29** L217
[24] Biémont E and Träbert E 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** 2939

[25] Berrington K A, Pelan J C and Waldock J A 2001 *J. Phys. B: At. Mol. Opt. Phys.* **34** L419
[26] Baudinet-Robinet Y, Garnir H-P, Dumont P-D and Résimont J 1990 *Phys. Rev.* A **42** 1080
[27] Curtis L J, Berry H G and Bromander J 1971 *Phys. Lett.* A **34** 169
[28] Träbert E 2000 *Phys. Scr.* **61** 257
[29] Träbert E 2002 *Phys. Scr.* T **100** 88
[30] Träbert E 2002 *Can. J. Phys.* **80** 1481
[31] Schmidt H T *et al* 1994 *Phys. Rev. Lett.* **72** 1616
[32] Träbert E, Gwinner G, Knystautas E J and Wolf A 2003 *Can. J. Phys.* **81** 941
[33] Crespo López-Urrutia J R, Beiersdorfer P, Savin D W and Widmann K 1998 *Phys. Rev.* A **58** 238
[34] Träbert E, Beiersdorfer P, Brown G V, Smith A J, Utter S B, Gu M F and Savin D W 1999 *Phys. Rev.* A **60** 2034
[35] Crespo López-Urrutia J R, Beiersdorfer P and Widmann K 2006 *Phys. Rev.* A **74** 012507
[36] Beiersdorfer P, Schweikhard L, Crespo López-Urrutia J and Widmann K 1996 *Rev. Sci. Instrum.* **67** 3818
[37] Träbert E, Beiersdorfer P, Gwinner G, Pinnington E H and Wolf A 2002 *Phys. Rev.* A **66** 052507
[38] Drake G W F 1971 *Phys. Rev.* A **3** 908
[39] Lin C D 1975 *PhD Thesis* Columbia University, New York
[40] Johnson W R, Plante D R and Sapirstein J 1995 *Advances of Atomic, Molecular and Optical Physics* vol 35 ed B Bederson and H Walther (San Diego: Academic) p 255
[41] Doerfert J, Träbert E, Wolf A, Schwalm D and Uwira O 1997 *Phys. Rev. Lett.* **78** 4355
[42] Chen M H, Cheng K T and Johnson W R 2001 *Phys. Rev.* A **64** 042507
[43] Träbert E and Curtis L J 2006 *Phys. Scr.* **74** C46
[44] Vilkas M J and Ishikawa Y 2003 *Phys. Rev.* A **68** 012503
[45] Beiersdorfer P, Träbert E and Pinnington E H 2003 *Astrophys. J.* **587** 836
[46] Brenner G, Crespo López-Urrutia J R, Harman Z, Mokler P H and Ullrich J 2007 *Phys. Rev.* A **75** 032504
[47] Träbert E 2004 *Astron. Astrophys.* **415** L39
[48] Träbert E, Saathoff G and Wolf A 2004 *Can. J. Phys.* **37** 945
[49] Träbert E, Reinhardt S, Hoffmann J and Wolf A 2006 *Can. J. Phys.* **39** 945
[50] Lapierre A *et al* 2005 *Phys. Rev. Lett.* **95** 183001
[51] Lapierre A *et al* 2006 *Phys. Rev.* A **73** 052507
[52] Draganić I, Crespo López-Urrutia J R, DuBois R, Fritzsche S, Shabaev V M, Soria Orts R, Tupitsyn I I, Zou Y and Ullrich J 2003 *Phys. Rev. Lett.* **91** 183001
[53] Johnson W R 2006 private communication
[54] Moehs D P, Bhatti M I and Church D A 2001 *Phys. Rev.* A **63** 032515
[55] Smith S J, Chutjian A and Lozano J A 2005 *Phys. Rev.* A **72** 062504
[56] Träbert E, Gwinner G, Wolf A, Knystautas E J, Garnir H-P and Tordoir X 2002 *J. Phys. B: At. Mol. Opt. Phys.* **35** 671
[57] Nicolaides C A 1977 *Phys. Lett.* A **63** 209
[58] Träbert E, Hoffmann J, Krantz C, Wolf A, Ishikawa Y and Santana J A 2009 *J. Phys. B: At. Mol. Opt. Phys.* **42** 025002
[59] Schippers S, Schmidt E W, Bernhardt D, Yu D, Müller A, Lestinsky M, Orlov D A, Grieser M, Repnow R and Wolf A 2007 *Phys. Rev. Lett.* **98** 033001
[60] Cheng K-T, Chen M H and Johnson W R 2008 *Phys. Rev.* A **77** 052504
[61] Epp S W *et al* 2007 *Phys. Rev. Lett.* **98** 183001